# VII.3 Spouted Bed Electrodes for Direct Utilization of Carbon in Fuel Cells

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## Objectives

- Explore the extension/application of spouted bed electrode (SBE) systems to direct carbon fuel cell (DCFC) applications.
- Simulate anticipated hydrodynamics of DCFCs.
- In a rectangular spouted vessel hydrodynamics apparatus (SVHA) with simulated mixtures containing carbon particles, explore fluid-particle circulation as a function of particle size, loading, fluid velocity, density, viscosity, geometry, etc.

## Approach

- Adapt and apply computational fluid dynamics (CFD) code originally developed to simulate particle circulation in spouted bed electrolytic reactors to carbon particle circulation in candidate geometries for DCFC systems.
- Experimentally investigate the hydrodynamics of carbon slurry circulation in DCFC systems using simulated slurry mixtures.

## Accomplishments

- A CFD hydrodynamics model of a rectangular spouted bed has been adapted to the DCFC application.
- Preliminary results involving particle loading and flow rate have been used to identify conditions for good particle contact with the inclined vessel bottom (e.g., anode), as well as particle recirculation.
- Both two-phase (liquid-solid) and three-phase (liquid-solid-$CO_2$ gas) hydrodynamics have been examined.
- The rectangular spouted vessel hydrodynamics apparatus has been retrofitted for simulated molten carbonate/carbon slurry experiments. Experiments are ongoing.

## Future Directions

- The CFD model will continue to be developed to explore the behavior of the spouted bed DCFC.
- The experimental apparatus will be used to conduct experiments using additives in water, simulate molten carbonate viscosity and density under expected DCFC operating conditions, and study carbon particles of varying sizes as a function of loading and flow rate.

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## Introduction

Carbon-oxygen fuel cells have been demonstrated that operate on very fine particles (10-1000 nm) of low-ash, turbostratic carbon. As impressive as these devices are, however, a number of problems remain to be solved before this technology can successfully operate on many different carbon sources on a large scale. The use of coal- and biomass-derived carbons, for example, requires a fuel cell design that can handle high amounts of ash and larger particles. It is proposed to

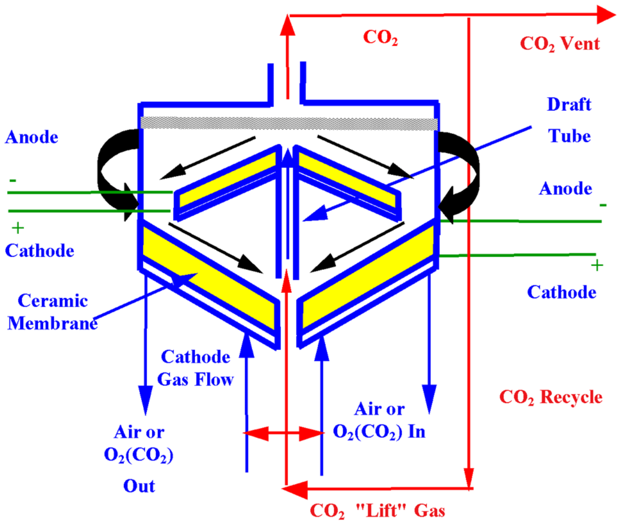


**Figure 1.** Schematic of Spouted Bed Direct Carbon Fuel Cell

adapt the concept of particulate spouted bed electrodes (SBEs) to the carbon-oxygen fuel cell.

In our laboratory, we have developed SBEs for point source metals recovery. In these devices, an aqueous solution containing metal ions is introduced as a high-velocity jet at the bottom of a conical vessel *via* a central draft tube. This liquid jet entrains particles that disengage from the jet in a region above the draft tube known as the "fountain." The particles are then distributed radially by the distributor to the bed periphery, where they fall onto the cathodic "feeder cone" at the reactor bottom and are directed back to the inlet of the draft tube for re-entrainment. Metal is deposited on the particles when they are in contact with the cathodic bottom cone. The "pumping action" of the spout continually circulates the particles through the vessel: upwards in the spout and downwards in the annular region. We have successfully recovered Ag, Au, Cu, Ni, Sn, and Cd from various aqueous solutions with this system at high current densities and high current efficiencies.

The objective of this project is to explore the extension/application of SBE-like systems to direct carbon fuel cells. More specifically, the approach is to (1) adapt SBE CFD codes to simulate the expected hydrodynamics of a DCFC and (2) perform simulated experiments of fluid-particle circulation in a rectangular spouted vessel hydrodynamics apparatus.

## Approach

A conceptual schematic of the spouted bed DCFC (SBDCFC) is presented in Figure 1. Carbon/molten carbonate slurry flows continuously through the system in a toroidal fashion – upwards in the draft tube and then centripetally back down along inclined, bipolar anode/cathode stacks. Two such stacks are shown in Figure 1 (one at the vessel bottom and the other serves as a slurry distributor); however, it is anticipated that a number of these stacks will be arranged in a single vessel in chevron-like arrangements.

The slurry can be circulated through the fuel cell either mechanically or by gas-lift created by injecting $CO_2$ produced in the carbon oxidation reaction into the slurry below the draft tube entrainment region (e.g., as in a gas-lift riser).

Fuel cell operation requires continuous carbon feed and removal of spent, ash-rich particles. The flow in the SBDCFC provides a few possibilities for accomplishing this. For example, the slurry can be circulated in an external circuit to remove spent, ash-rich particles (e.g., by filtration with metallic or ceramic media), and to feed fresh carbon particles to the SBDCFC. These operations can also be performed within the cell either at the draft tube inlet or outlet.

In order to become a viable system, the SBDCFC must be capable of operation on a large scale. The SBDCFC has some significant advantages in this regard: (1) it is readily scalable (e.g., by "stacking" units and inclined bipolar elements); and (2) continuous flow provides the means for performing removal and recovery of spent particles, continuous "self-cleaning" of electrodes that can be easily fouled in other systems, and facile temperature control *via* convective heat removal. In particular, the "self-cleaning" nature of the electrodes tends to minimize the effects of contaminants on performance and aging.
In addition, the continuous feed of carbonaceous material tends to reduce electrode sintering that can be a problem for fixed solid electrodes.

As a result of our spouted bed electrolytic reactor work, we have developed numerical Eulerian CFD

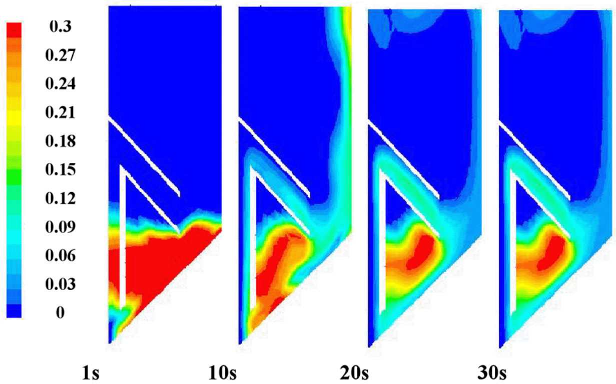


**Figure 2.** Simulation Results Using an Eulerian CFD Model Solved with FLUENT™ of Time Evolution of Carbon Particle Volume Fraction in One Quadrant of a Rectangular Spouted Vessel; $Li_2(CO_3)$ at 810°C; 200 g, 0.1 mm Particles; 0.3 m/s Inlet Velocity

models for simulating the performance of SBE systems. For electrowinning, these models were validated with hydrodynamic data from a rectangular SVHA. We are pursuing a similar approach for the SBDCFC concept, i.e., simulation of carbon slurry flows with the CFD model, and model validation *via* hydrodynamics experiments with simulated carbon/ molten carbonate slurries in the SVHA.

## Results

**Simulations.** A number of carbon/molten carbonate slurry flow conditions have been simulated in a domain of the same size as one quadrant of the SVHA experimental system. An example of these results is presented in Figure 2. The geometry in this case includes a draft tube with a "cap." Molten carbonate is injected at the inlet at 0.3 m/s. As shown, even though the densities of the carbon particles and molten carbonate are practically the same, under these conditions there is good contact of the carbon with the inclined vessel bottom, as well as the top and bottom of the distribution channel; all these inclined surfaces can potentially serve as anodes in a SBDCFC "chevron-type" bipolar electrode system.

Another simulation result is presented in Figure 3 with just an open draft tube in which $CO_2$ is used as a lift-gas to circulate the carbon/molten carbonate slurry in the SBDCFC. Two things are apparent in

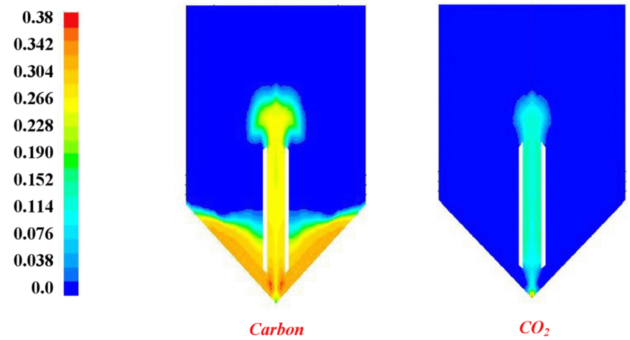


**Figure 3.** Simulation Results Using an Eulerian CFD Model Solved with FLUENT™ of Carbon Particle and $CO_2$ Gas Volume Fractions in Molten Carbonate at 29 s after Start-up with Pure $CO_2$ at Atmospheric Pressure at the Inlet, in a Rectangular SBDCFC; $Li_2CO_3$ at 810°C; 200 g, 0.1 mm Particles

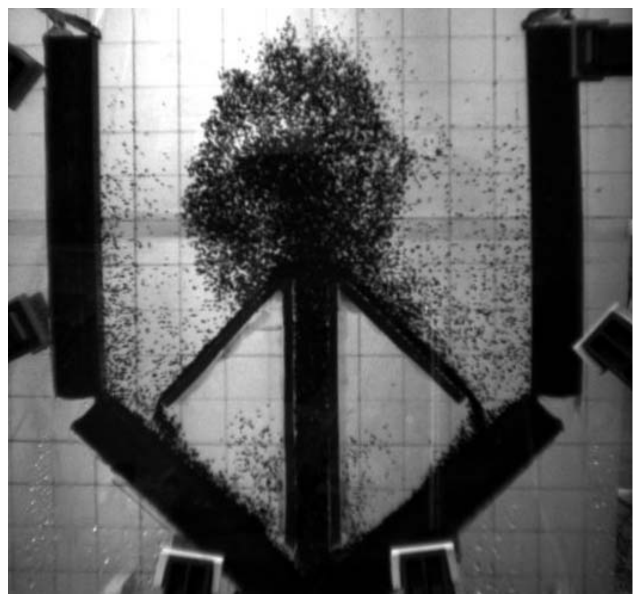

**Figure 4.** Carbon-Water Slurry Flow in SVHA; 2.6 gpm Water, 75 $cm^3$, 1 mm Carbon Particles

this simulation: (1) $CO_2$ injection does provide "lift" of the slurry through the draft tube; and (2) the transient start-up process is much slower than in Figure 2, where molten carbonate is being injected directly into the cell. This is undoubtedly due to the fact that the gas provides less force than the injected fluid, and it takes longer to develop the momentum required to circulate the slurry. Since this is a more complex, three-phase calculation, the solution diverges at about 29 s, so we have yet to determine the eventual steady-state condition.

**Experimental.** Simulated carbon slurry experiments are being conducted in the SVHA. An image of the apparatus is presented in Figure 4. A 5-hole Pitot probe is used to determine the velocity field at selected points within the vessel. Fluid profile properties will be varied with additives to simulate carbon/molten carbonate slurry flow.

## Conclusions

- Hydrodynamics can be used to control the circulation, residence time, and distribution of carbon within the spouted bed.
- Function of relative densities, viscosity, particle loading and particle size.
- Carbonate slurry circulation can be provided by pumping the slurry or by gas-lift.
- Gas-lift more favorable – no moving parts.
- Good carbon particle contact with inclined anodes can be achieved with low inlet velocities and with larger particles with greater relative density.

## FY 2004 Publications/Presentations

*Conference Presentations*

1. "Hydrodynamic Reaction Model of a Spouted Bed Electrolytic Reactor," P.A. Shirvanian, and J.M. Calo, Paper No. 190a, presented at the AIChE 2003 Annual Meeting, San Francisco, CA, November 17, 2003.

2. "A Kinetic-theory Analysis of the Scale-up for the Hydrodynamics of a Rectangular Slot, Spouted Fluidized Vessel," P.A. Shirvanian and J.M. Calo, Paper No. 292g, presented at the AIChE 2003 Annual Meeting, San Francisco, CA, November 17, 2003.

3. "Spouted Bed Electrodes (SBE) For Direct Utilization of Carbon in Fuel Cells," J.M. Calo, DOE NETL UCR/HBCU Contractors' Review Meeting, Pittsburgh, PA, June 9-10, 2004.

*Papers*

1. "An Experimental Investigation of the Hydrodynamics of a Rectangular, Spouted Vessel with a Draft Duct," P.A. Shirvanian, J.M. Calo, and G. Hradil, submitted to *Chem. Eng. Sci.,* 2004.

2. "Numerical Simulation of Fluid-Particle Hydrodynamics in a Rectangular Spouted Vessel," P.A. Shirvanian, J.M. Calo, and G. Hradil, in press, *Int. J. Multiphase Flow,* 2004.

3. "Hydrodynamic Scaling of a Rectangular Spouted Vessel With a Draft Duct," P.A. Shirvanian and J.M. Calo, submitted to *Chem. Eng. J.,* 2003.